

Chemical and size effects of hygroscopic aerosols on light scattering coefficients

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Abstract. The extensive thermodynamic and optical properties recently reported [Tang and Munkelwitz, 1994a] for sulfate and nitrate solution droplets are incorporated into a visibility model for computing light scattering by hygroscopic aerosols. The following aerosol systems are considered: NH_4HSO_4 , $(\text{NH}_4)_2\text{SO}_4$, $(\text{NH}_4)_3\text{H}(\text{SO}_4)_2$, NaHSO_4 , Na_2SO_4 , NH_4NO_3 , and NaNO_3 . In addition, H_2SO_4 and NaCl are included to represent freshly formed sulfate and background sea-salt aerosols, respectively. Scattering coefficients, based on $1\text{ }\mu\text{g}$ dry salt per cubic meter of air, are calculated as a function of relative humidity for aerosols of various chemical compositions and lognormal size distributions. For a given size distribution the light scattered by aerosol particles per unit dry-salt mass concentration is only weakly dependent on chemical constituents of the hygroscopic sulfate and nitrate aerosols. Sulfuric acid and sodium chloride aerosols, however, are exceptions and scatter light more efficiently than all other inorganic salt aerosols considered in this study. Both internal and external mixtures exhibit similar light-scattering properties. Thus for common sulfate and nitrate aerosols, since the chemical effect is outweighed by the size effect, it follows that observed light scattering by the ambient aerosol can be approximated, within practical measurement uncertainties, by assuming the aerosol being an external mixture. This has a definite advantage for either visibility degradation or climatic impact modeling calculations, because relevant data are now available for external mixtures but only very scarce for internal mixtures.

Introduction

Inorganic sulfates and nitrates frequently constitute a major fraction of the ambient aerosol. In particular, atmospheric sulfates are often found to be associated with the fine particulate mass, in the size range between 0.1 and $1.0\text{ }\mu\text{m}$, where a particle scatters sunlight most efficiently. Thus a strong correlation between visibility degradation and sulfate mass concentration has always been observed [Waggoner *et al.*, 1976; White, 1976; Leaderer *et al.*, 1979]. In more detailed studies, investigators have applied multiple regression analyses to apportioning light scattering and absorption to all gaseous species and chemical constituents in ambient aerosols measured at various locations [White and Roberts, 1977; Cass, 1979; Hasan and Dzubay, 1983; Malm *et al.*, 1994]. Visibility models have also been developed to compute light extinction coefficients from the Mie theory, using either the measured or the estimated physical properties of the aerosol and the known optical and thermodynamic properties of its constituents [Tang *et al.*, 1981; Sloane, 1983; Sloane and Wolff, 1985; Larson *et al.*, 1988].

Inorganic salt aerosols are mostly hygroscopic by nature and exhibit the property of deliquescence in humid air [Tang, 1980]. The phase transformation from a solid particle to a saline droplet usually occurs spontaneously when the relative humidity in the surrounding atmosphere reaches a level, known as the deliquescence point, that is specific to the chemical composition of the aerosol particle [Orr *et al.*, 1958; Tang, 1976; Tang and Munkelwitz, 1993]. A typical growth curve for a

$(\text{NH}_4)_2\text{SO}_4$ particle upon increasing humidity is shown by open circles in Figure 1, where the particle mass change by either acquiring or losing water is plotted as a function of the relative humidity (%RH). The phase transformation at the deliquescence point, 80% RH for $(\text{NH}_4)_2\text{SO}_4$, is clearly indicated. The droplet growth above the deliquescence point can be computed from the water activity data available in the literature for solutions below the saturation concentration. Upon decreasing humidity, the droplet loses water by evaporation and continues to do so gradually even below the saturation (or deliquescence) point, as shown by the solid circles in Figure 1, until about 38% RH for $(\text{NH}_4)_2\text{SO}_4$ when suddenly it crystallizes. The supersaturated solution region between the saturation and the crystallization points is a metastable state readily attained by suspended saline droplets but not by the bulk solution. Field measurements by Rood *et al.* [1989] have revealed that such supersaturated solution droplets are indeed present abundantly and ubiquitously in the atmosphere. Until very recently, however, few data are available for visibility modeling over the low relative humidity region where atmospheric aerosol particles are expected to be present in the form of supersaturated solution droplets.

In this paper the extensive thermodynamic and optical properties reported by Tang and Munkelwitz [1994a] for sulfate and nitrate solution droplets are incorporated into a visibility model for computing light scattering by hygroscopic aerosols much below the deliquescence humidities. The aerosol systems considered include NH_4HSO_4 , $(\text{NH}_4)_2\text{SO}_4$, $(\text{NH}_4)_3\text{H}(\text{SO}_4)_2$, NaHSO_4 , Na_2SO_4 , NH_4NO_3 , and NaNO_3 . In addition, H_2SO_4 and NaCl are also included in the study to represent freshly formed sulfate and background sea-salt aerosols, respectively. The effects of aerosol size, size distribution, chemical compo-

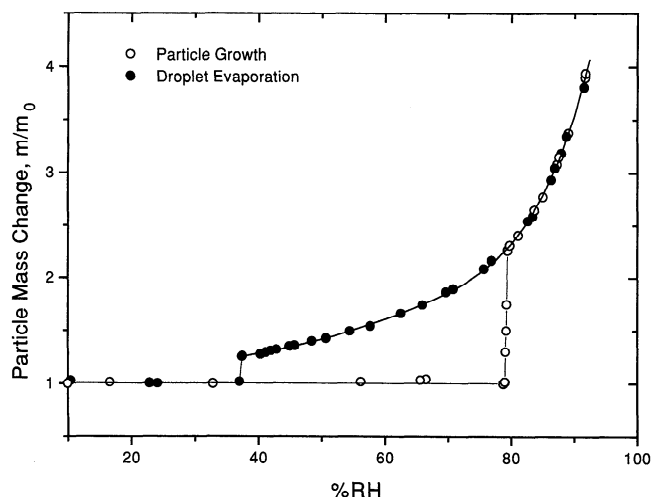


Figure 1. Phase transformation, growth, and evaporation of an $(\text{NH}_4)_2\text{SO}_4$ particle as a function of relative humidity at 25°C .

sition, and relative humidity on light scattering are studied by model computations. Contributions by external and internal mixtures are also compared and discussed.

Scattering Coefficient Calculations

The scattering coefficient, b_{sca} , for an aerosol of given size distribution, $f(D)$, with respect to diameter D may be computed, using the following formula:

$$b_{\text{sca}} = \int_0^\infty \pi(D/2)^2 Q_{\text{sca}}(\alpha, n) N f(D) dD \quad (1)$$

where α , the optical parameter, is equal to $\pi D/\lambda$. N is particle number concentration, and $Q_{\text{sca}}(\alpha, n)$ is single-particle scattering cross section for given α , wavelength λ , and refractive index n . The wavelength, $\lambda = 0.58 \mu\text{m}$, is chosen as recommended by *Presle and Horvath* [1978] to give the maximum perception of an object under daylight conditions. At this wavelength there is negligible absorption by either salts or their aqueous solutions and therefore b_{sca} is equal to the extinction coefficient b_{ext} .

Throughout this study, a constant dry-salt loading, w_0 , of $1 \mu\text{g m}^{-3}$ of air was employed as the basis of computation. Thus for a lognormal size distribution of dry-salt particles with count median diameter D_g and geometric standard deviation σ_g the particle number concentration N is computed from the mass balance:

$$\begin{aligned} w_0 &= (4/3) \pi \rho_0 N \int_0^\infty (D_0/2)^3 f(D_0) dD_0 \\ &= (1/6) \pi \rho_0 N D_g^3 \exp(4.5 \ln^2 \sigma_g) \end{aligned} \quad (2)$$

where D_0 is the diameter of the dry salt of density ρ_0 . *Willeke and Brockmann* [1977] have made extensive computations of extinction coefficients for lognormally modeled, multimodal particle volume distributions without considering particle growth. The particle number distribution chosen in this study facilitates light-scattering computations for particles undergo-

ing growth and evaporation in a changing humidity environment, which is the main interest of this study.

The droplet growth factor, β , which is the change in size of a solution droplet in equilibrium with the surrounding humidity, is defined by the following equation:

$$\beta = D/D_0 = \left(\frac{100}{x} \frac{\rho_0}{\rho} \right)^{1/3} \quad (3)$$

where D is the diameter of the droplet of density ρ at x wt %. For droplets the equilibrium relative humidity is increased by the Kelvin effect due to the surface tension of the droplet curvature according to the following equation:

$$\% \text{RH} = 100 a_w \exp \left(\frac{4 \gamma M}{\rho R T D} \right) \quad (4)$$

where a_w and γ are, respectively, the water activity and surface tension of a bulk solution of composition x and density ρ . M is the molecular weight of water, R the gas constant, and T the absolute temperature. For a typical solution droplet, say $(\text{NH}_4)_2\text{SO}_4$, of diameter $0.1 \mu\text{m}$, the exponential factor amounts to 1.017, using $\gamma = 82.3 \text{ dyn cm}^{-2}$ and $\rho = 1.394 \text{ g cm}^{-3}$ for a 40 wt % solution. It means that the curvature effect would raise the equilibrium relative humidity of this droplet by only 1.7% above what would be required for a bulk solution of the same composition. Since surface tension data are not readily available for supersaturated solutions, the curvature effect is generally neglected in droplet growth/evaporation computations for aerosol particles in the optical size range. Thus throughout this study, % RH is given by $100 a_w$.

The density ρ and water activity a_w are expressed by polynomials:

$$\rho = 0.9971 + \sum A_i x^i \quad (5)$$

$$a_w = 1.0 + \sum C_i x^i \quad (6)$$

Except for NaCl , H_2SO_4 , and NH_4NO_3 the coefficients A_i and C_i were taken from *Tang and Munkelwitz* [1994a]. Refractive indices were computed from the appropriate partial molal refraction values, R_1 and R_2 , also given by *Tang and Munkelwitz* [1994a], using the formula

$$R = y_1 R_1 + y_2 R_2 = V(n^2 - 1)/(n^2 + 2) \quad (7)$$

where y and M are mole fraction and molecular weight, respectively, and subscripts 1 and 2 refer to water and the solute, respectively. V is the molar volume given by

$$V = \frac{1}{\rho} (y_1 M_1 + y_2 M_2) \quad (8)$$

Data for aqueous NaCl , H_2SO_4 , and NH_4NO_3 solutions at 25°C are given in Table 1. For dry salts the density ρ_0 and refractive index n_0 values are listed in Table 2, together with the relative humidities at respective deliquescence points (% RHD) and crystallization points (% RHC).

Scattering coefficient calculations were performed on a 486-based PC, following essentially the algorithm given by *Tang et al.* [1981]. (Note that in the 1981 paper the numerical integration steps at each RH were inadvertently multiplied by a droplet growth factor, making the computed extinction coefficients larger by such a factor. This error has since been corrected in later calculations.)

Table 1. Thermodynamic and Optical Properties of H_2SO_4 , NH_4NO_3 and NaCl Solutions at 25°C

	H_2SO_4	NH_4NO_3	NaCl
C_1	-5.196 (-3)	-3.65 (-3)	-6.366 (-3)
C_2	9.746 (-5)	-9.155 (-6)	8.624 (-5)
C_3	-9.693 (-6)	-2.826 (-7)	-1.158 (-5)
C_4	9.405 (-8)	0	1.518 (-7)
A_1	7.367 (-3)	4.05 (-3)	7.41 (-3)
A_2	-4.934 (-5)	9.0 (-6)	-3.741 (-5)
A_3	1.754 (-6)	0	2.252 (-6)
A_4	-1.104 (-8)	0	-2.06 (-8)
R_2	13.45	15.08	8.95

Results and Discussions

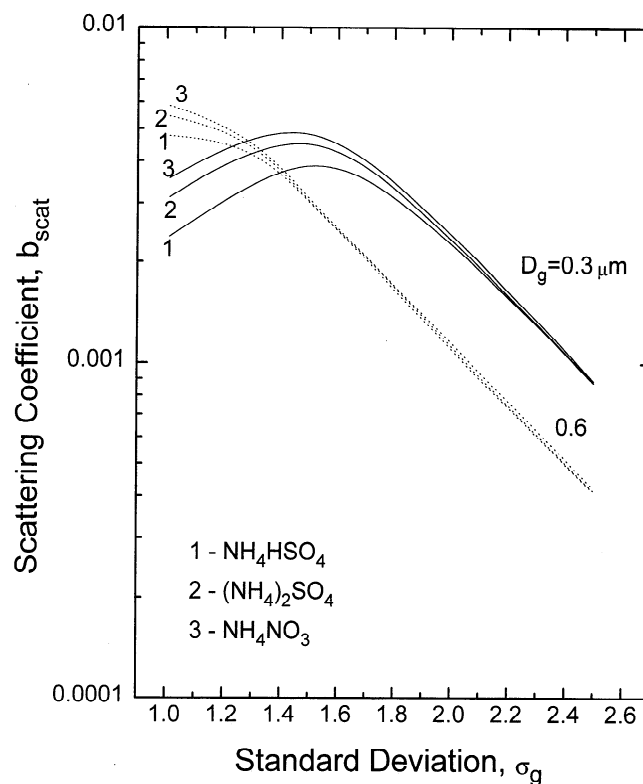
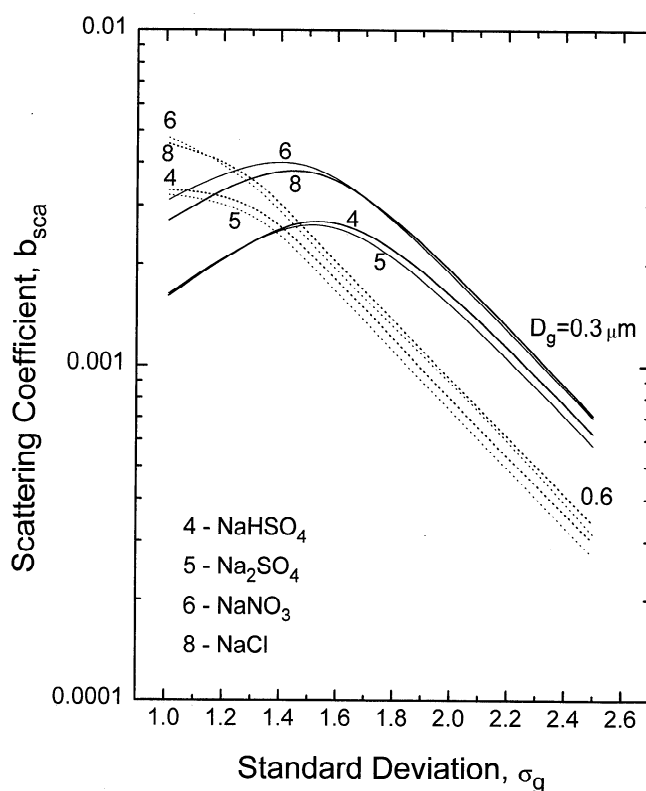
Dry-salt aerosols. Scattering coefficients, b_{sca} ($\text{km}^{-1}/\mu\text{g}$ dry salt m^{-3}), were first computed for dry-salt aerosols of two diameters, $D_g = 0.3$ and $0.6 \mu\text{m}$, as a function of σ_g varying from 1.01 to 2.5, using the data in Table 2. All dry-salt particles are assumed to be spherical in shape so that Mie computations can be performed. The results are presented in Figures 2 and 3 for NH_4^+ and Na^+ salts, respectively. The NH_4^+ -salt aerosols exhibit similar light-scattering properties, especially at broader size distributions, because these salts have similar refractive indices and densities. On the other hand, the Na^+ -salt aerosols scatter light less efficiently than corresponding NH_4^+ -salt aerosols for the same size distribution. This is so because the Na^+ salts have larger densities and therefore according to (2) produce fewer particles as scatterers per unit mass concentration.

Droplet growth factors. The droplet growth factor over the dry particle, β , may be computed from (3) for each electrolyte at any relative humidity, as long as the particle remains a solution droplet. This factor is species specific and independent of the size distribution of an aerosol. Figure 4 shows the computed droplet growth factors as a function of % RH for all electrolytes considered in this study. It is interesting to note that droplets containing Na^+ salts (dashed curves, excluding NaCl) appear to grow larger than the NH_4^+ -salt solution droplets (solid curves). Both H_2SO_4 and NaCl solution droplets grow even larger than all other salt solution droplets at $\sim 80\%$ RH or lower. Furthermore, the growth of the NH_4^+ -salt solution droplets below $\sim 90\%$ RH tends to differ very little from one another. At 80% RH the droplet growth factors of the NH_4^+ -salt aerosols have a value close to 1.5, a value quoted by Hegg *et al.* [1993] in their discussion on the effect of relative humidity on light scattering by tropospheric aerosols.

Hydrated aerosols. Light scattering by aerosols composed of saline droplets depends, in principle, on both optical and

Table 2. Thermodynamic and Optical Properties of Dry-Salt Aerosols

	Density	Refractive Index	% RHD	% RHC
$\text{NH}_4\text{H SO}_4$	1.78	1.473	40	20-0.05
$(\text{NH}_4)_3\text{H}(\text{SO}_4)_2$	1.83	1.51	69	44-35
$(\text{NH}_4)_2\text{SO}_4$	1.76	1.53	80	40-37
NH_4NO_3	1.725	1.554	62	25-32
NaHSO_4	2.476	1.46	52	<0.05
Na_2SO_4	2.68	1.48	84	59-57
NaNO_3	2.261	1.587	74.5	30-0.05
NaCl	2.165	1.544	75.3	48-46

**Figure 2.** Light scattering by dry NH_4^+ -salt aerosols.**Figure 3.** Light scattering by dry Na^+ -salt aerosols.

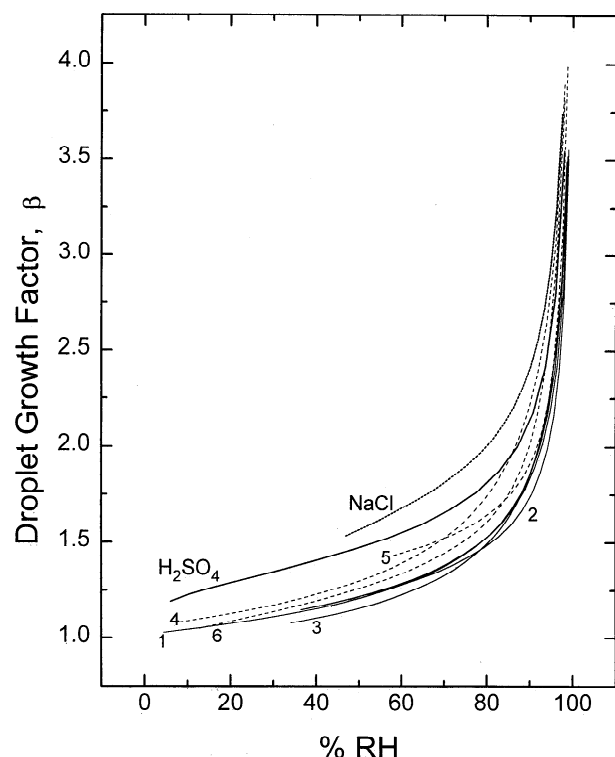


Figure 4. Droplet growth factors for hygroscopic aerosols: 1, NH_4HSO_4 ; 2, $(\text{NH}_4)_2\text{SO}_4$; 3, NH_4NO_3 ; 4, NaHSO_4 ; 5, Na_2SO_4 ; 6, NaNO_3 ; 7, $(\text{NH}_4)_3\text{H}(\text{SO}_4)_2$. Number 7 lies between 1 and 2 and is not clearly visible.

chemical properties of the aerosol. Scattering coefficients, b_{sca} ($\text{km}^{-1}/\mu\text{g dry salt m}^{-3}$), were computed as a function of % RH for the hygroscopic aerosols of two dry-salt size distributions: $(D_g, \sigma_g) = (0.3 \mu\text{m}, 1.5)$ and $(0.6 \mu\text{m}, 1.5)$. The results are presented in Figures 5 and 6, respectively. Here, to illustrate particle growth/evaporation but not overcrowd the plots, light-scattering curves of a complete growth/evaporation cycle are shown for $(\text{NH}_4)_2\text{SO}_4$ and NaNO_3 aerosols only. All other curves represent droplet evaporation down to approximately the crystallization relative humidities specific to each species. The results show that similar to the dry-salt aerosols discussed above, hygroscopic aerosols of the larger size ($0.6 \mu\text{m}, 1.5$) scatter light less efficiently than the smaller size ($0.3 \mu\text{m}, 1.5$) aerosols even after they have grown into droplets. Except for H_2SO_4 and NaCl , all hygroscopic aerosols of the same initial dry-salt size distribution appear to group together into a rather narrow band, indicating a very weak dependence on the chemical constituent of the aerosol. In addition, NH_4^+ salts (solid lines) and Na^+ salts (dashed lines) seem to form separate subgroups within the band. Both H_2SO_4 and NaCl aerosols, however, scatter light more efficiently than all other sulfate and nitrate aerosols considered in this study.

Hegg *et al.* [1993] have discussed a hygroscopic growth factor (not to be confused with the droplet growth factor discussed above) for tropospheric aerosols, which is defined as the ratio of the light scattering by an aerosol at 80% RH to that at 30% RH. At 30% RH, most of the hygroscopic aerosols are dehydrated, except for a few highly hygroscopic salts such as NH_4HSO_4 and NaHSO_4 [Tang and Munkelwitz, 1994a]. This hygroscopic growth factor has been identified as a key parameter in the assessment of the climatic impact of sulfate aerosols

[Charlson *et al.*, 1992]. A single global value of 1.7 ± 0.3 , derived from direct nephelometer measurements at a few selected geographical locations, has been utilized to date as a first estimate in climate change modeling studies. Hegg *et al.* [1993], in pointing out the theoretical basis of this growth factor, have showed that the factor is influenced by the position of the initial dry aerosol size distribution relative to the efficient light-scattering droplet size range. For the two initial dry-salt size distributions studied in this work, namely, $(0.3 \mu\text{m}, 1.5)$ and $(0.6 \mu\text{m}, 1.5)$, the hygroscopic growth factor for the NH_4^+ -salt aerosols has a mean value of approximately 2.41 and 2.13, respectively. The factor becomes even larger with smaller particle size and narrower size distribution, as also pointed out by Hegg *et al.* [1993]. On the other hand, if the aerosol particles are not completely dry at 30% RH, as in the case of NH_4HSO_4 , the two size distributions studied here would yield respective hygroscopic growth factors of 2.16 and 1.76, which are closer to the observed global value of 1.7 ± 0.3 .

Mixed-salt aerosols. Since the ambient aerosol is far from being a single-salt aerosol, the question of external and internal mixtures often comes up when assessing the contributions to light extinction by chemical components in the aerosol. The findings of this work show that for common hygroscopic aerosols the chemical effect is rather outweighed by the size effect. It follows that observed light scattering may be approximated without appreciable errors by assuming the aerosol being an external mixture. An example is given here for the mixed-salt aerosol composed of 55.5% by wt NH_4NO_3 and 44.5% $(\text{NH}_4)_2\text{SO}_4$. The optical and thermodynamic data for this mixed-salt system have been reported by Tang *et al.* [1981]. The results are presented in Figure 7, where light-scattering coef-

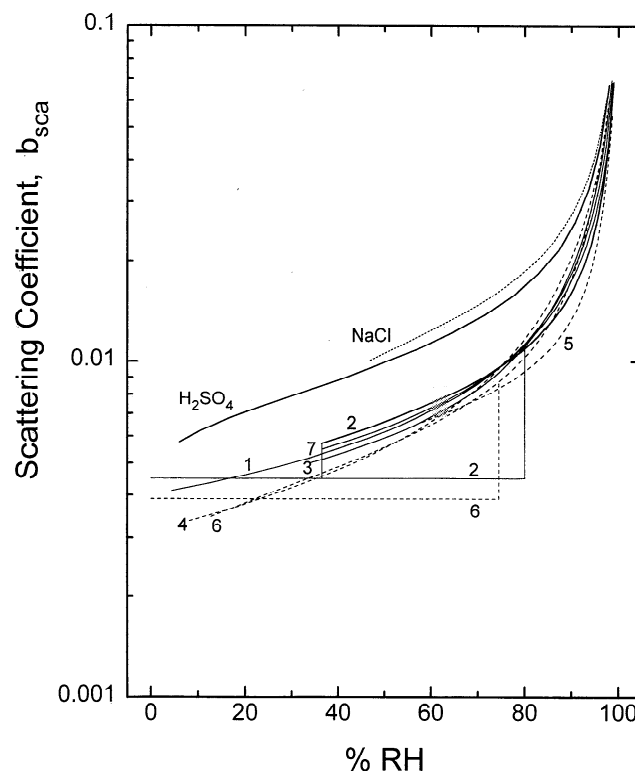


Figure 5. Light scattering by hygroscopic aerosols ($D_g = 0.3 \mu\text{m}$, $\sigma_g = 1.5$). 1, NH_4HSO_4 ; 2, $(\text{NH}_4)_2\text{SO}_4$; 3, NH_4NO_3 ; 4, NaHSO_4 ; 5, Na_2SO_4 ; 6, NaNO_3 ; 7, $(\text{NH}_4)_3\text{H}(\text{SO}_4)_2$.

ficients are plotted as a function of % RH for both single-salt aerosols and external and internal mixtures of two dry-salt size distributions: ($0.3\ \mu\text{m}$, 1.5) and ($0.6\ \mu\text{m}$, 1.5). As expected, the light-scattering curves for each size distribution fall inside a narrow band, indicating that within practical measurement uncertainties the chemical effect is negligible. Note that, here, the aerosol particles are assumed to remain as solution droplets in the relative humidity range considered. If, however, atmospheric conditions prevail such that the relative humidity has fallen below the crystallization points and that some aerosol particles are still in the solid state, light scattering by such a multicomponent and multiphase aerosol certainly requires due consideration of the deliquescence properties of hygroscopic aerosols [Tang and Munkelwitz, 1993, 1994b].

Conclusions

Light-scattering coefficients as a function of % RH have been calculated for the sulfate and nitrate aerosols of atmospheric importance, using the extensive optical and thermodynamic properties recently published. It is shown that the chemical effect on light scattering is outweighed by the size effect of the aerosols under consideration. The contributions by external and internal mixtures are compared and found to differ only slightly. It is concluded that at least for the common sulfate and nitrate salt aerosols, external mixtures may be used as an aerosol surrogate in model computations to predict light-scattering properties. This has a definite advantage for either visibility degradation or climatic impact modeling studies, since relevant data are now available for pure components but only very scarce for internal mixtures. This work also points out the

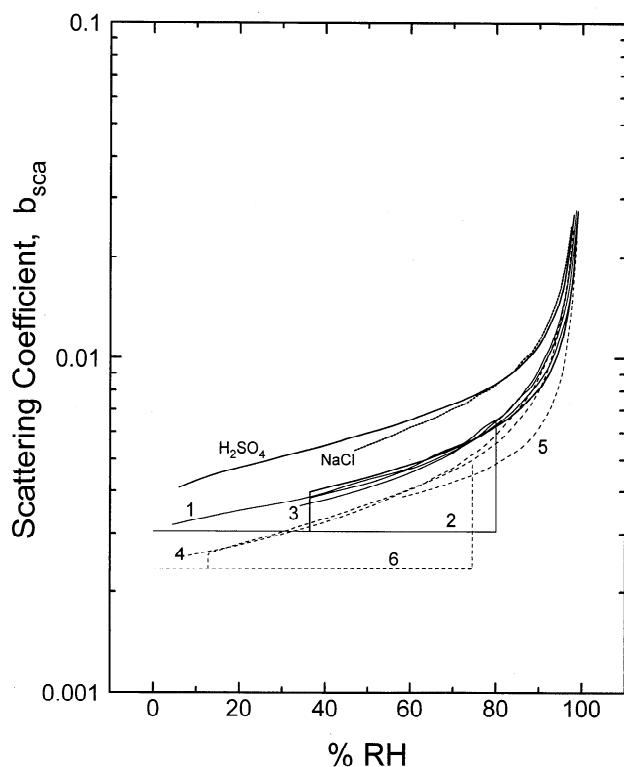


Figure 6. Light scattering by hygroscopic aerosols ($D_g = 0.6\ \mu\text{m}$, $\sigma_g = 1.5$): 1, NH_4HSO_4 ; 2, $(\text{NH}_4)_2\text{SO}_4$; 3, NH_4NO_3 ; 4, NaHSO_4 ; 5, Na_2SO_4 ; 6, NaNO_3 ; 7, $(\text{NH}_4)_3\text{H}(\text{SO}_4)_2$. Number 7 lies between 1 and 2 and is not clearly visible.

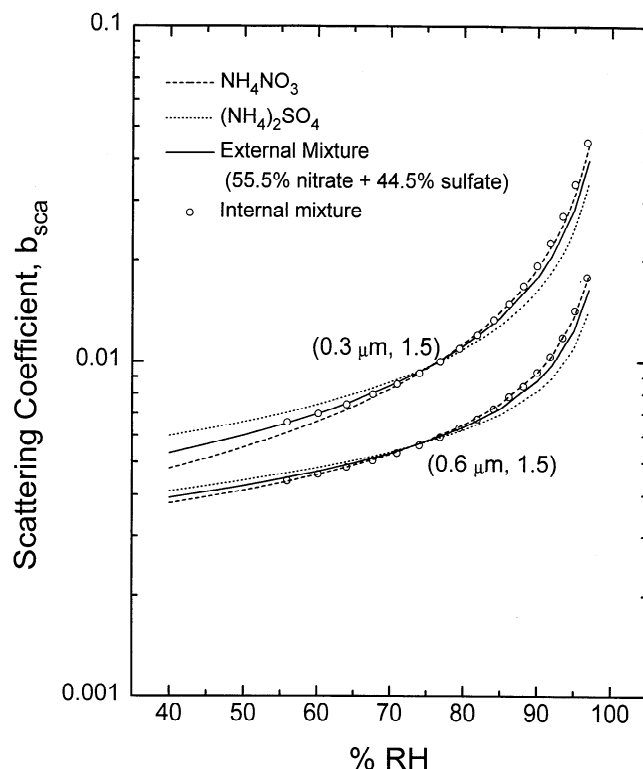


Figure 7. Light scattering by mixed-salt aerosols: external versus internal mixtures.

need for more information on the optical and thermodynamic properties of the highly acidic aerosols and background sea-salt aerosols, as these aerosols are shown to fall outside the bounds for the common sulfate and nitrate aerosols in light-scattering behavior.

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